SPUTTERING FROM OBLIQUE ION INCIDENCE USING COMPUTER SIMULATION TECHNIQUE

Horace Truman Holcombe

United States
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ABSTRACT

A self-contained computer simulation of sputtering from 20 keV incident ions is not possible with present computers. However, a simulation can be done by considering primary and secondary collisions separately. An investigation of 20 keV argon ions incident obliquely on the (100) surface of a face-centered cubic copper crystal was done at angles from 29 to 61 degrees from normal. Results strongly support the concept of transparency, but indicate that focused collision sequences make very limited contributions to sputtering. Depending on the ion beam incidence angle, up to 25 percent of the sputtering may be due to random collision cascades initiated by deep primary collisions. The remainder is caused by surface collision mechanisms. Reflection of incident ions off surface atoms significantly affects argon-copper sputtering when the ions are obliquely incident.

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ACKNOWLEDGEMENTS

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I am especially grateful to Professor D.E. Harrison, Jr., for his stimulating discussion, assistance, and encouragement while doing the research for this project and writing the manuscript.

I. INTRODUCTION

When a beam of ions is directed toward a solid surface, atoms are removed by momentum reversal processes. This phenomenon is known as sputtering. It has been observed for over a century, but only recently have experimental techniques been refined sufficiently to insure that results are reproducible. Likewise, only recently have serious attempts been made to explain the details of the momentum reversal theoretically.

The most interesting results in recent years have been obtained from investigations of ion bombardment of single crystals. Wehner [1] was first to observe that atoms sputtered from a single crystal move in preferred directions, and that this effect reflects characteristics of the bombarded surface. The experimental results were convincing evidence that momentum reversal was due to collision processes and dependent on the geometry of the crystal. Shortly thereafter Silsbee [2] showed, using classical two-body collision theory, that under certain conditions (the atom under consideration has energy below some threshold, E_f) the angle between the axis of a close packed row in a lattice and the direction of motion of an atom in the row will decrease in subsequent collisions. This results in "focusing" of momentum along close-packed rows. Numerous additions and refinements have been made [3,4,5], and many investigators have concluded that focusing mechanisms are responsible for the observed anisotropic sputtering from single crystals.

Nelson and Thompson [4] carried out experiments particularly designed to illustrate focusing as the mechanism which produces the observed sputter patterns. They introduced a second type of focusing mechanism to account for patterns experimentally observed which could not be explained by the simple Silsbee sequence. This mechanism does not rely solely on a collision, but introduces the concept of focusing action on a moving atom by surrounding rings of atoms in a lattice. Again this "assisted" focusing sequence propagates along rows of atoms only if the energy is below a focusing threshold, E_f. Thompson [5] has considered a further mechanism of importance for high energy (40 keV) bombardment. He suggested that atomic collision cascades initiated by the bombarding ions generates focused collision sequences when the atom energies fall below E_f. He concluded, from bombardment of a gold monocrystal by singly ionized xenon and argon, that random cascades and focused collision sequences contribute approximately equal amounts to the total sputtering yield.

However Harrison, Levy, Johnson, and Effron [7] have shown, using a computer simulation technique similar to that of Gibson, Goland, Milgram, and Vineyard [8], that collisions, which give rise to patterns of sputtered atoms that agree quite closely with experiment, occur at or near the surface. Likewise, Lehman and Sigmund [9] have shown that much of the data previously assumed to indicate the presence of focusing can be described by simple

collisions with a symmetric array. Further Harrison [10] has done computer studies, simulating argon ions incident on a (110) surface of copper, which indicate that only a small part of the incident beam cross-section could cause collisions of the type which lead to focusing mechanisms.

Fluit and Rol [6] have done experiments on copper monocrystals demonstrating conclusively the effect of the periodic nature of the atomic structure on the sputtering process. Their results do not necessarily support the idea that focusing mechanisms are important in sputtering, but simply demonstrate the following: When the copper crystal is aligned so that incoming ions see relatively open areas (channels) between the atomic rows, the ions penetrate to such depths that a collision sequence resulting in surface damage is unlikely. Minima on a plot of sputtering ratio versus angle of beam incidence correspond approximately to the most "transparent" directions as determined by the geometry of the crystal.

II. OBJECTIVES

While there is no doubt that focused collision sequences occur in crystalline solids, they have not previously been observed to cause significant sputtering in computer simulations. In such simulations focused collision sequences are initiated in random cascades and follow close packed rows of atoms, as experimentally observed by Thompson [5]. In crystal orientations previously studied by computer simulations [7,8,10], the close packed row axes are oriented either parallel or 45 degrees to the bombarded surface. Thus for a collision sequence initiated several atomic layers below the surface, the resultant sputtering will occur relatively far away from the impact point of an incoming ion. Present computers do not have enough memory capacity or speed to simulate a crystal large enough to contain all such events from a high energy (20 keV or greater) incident ion.

The first objective of this study was simply to find a collision sequence resulting in sputtering in a simulated ion-lattice collision, and to determine the conditions under which it occured. The second and primary objective was to determine the importance of focused collision sequences in the sputtering process in general.

III. SIMULATION MODELS AND PROCEDURES

The sputtering simulation computer programs used in this work were developed by Harrison, Levy, Johnson, and Effron [7] and may be generally described as follows: A space lattice is defined whose atomic sites are those of a face-centered cubic copper monocrystal. The displacement of each atom is then computed by stepwise numerical integration of its equations of motion as an energetic argon ion collides with the lattice. Computation continues until all significant collisions within the lattice are complete.

A. THE LATTICE MODEL

The lattice model used is with minor modifications the same as that used in a number of previous works [7,10,12,13]. Effron, Gay, and Harrison [11] give a detailed description of the collision dynamics and theoretical justification for the model which need not be repeated here.

As before, the interatomic potential describing copper-copper interactions is the Born-Mayer type Gibson Number Two [8] which is eroded at half the nearest neighbor distance so that the undisturbed lattice is stable. Copper-argon interactions are governed by the Born-Mayer potential (KSE-B) determined by Harrison, Carlston, and Magnuson [14] from a study of secondary electron emission.

In a recent study by Harrison and Moore [15] a lattice model which included an attractive (Morse) term in the copper-copper interaction potential was used in a simulation program. Using parameters calculated by Girifalco and Weizer [16], it was found that:

(1) The surface layers relaxed outward slightly to a new equilibrium position.

(2) The Morse potential produced a negligible difference in the collision dynamics.

(3) The program required much more running time than those with only repulsive potentials.

This model produced essentially the same sputtering as the original lattice model with the artificial addition of a sputtering energy threshold (as had already been done [7]). This threshold, associated with only the velocity component normal to the surface, was found to be approximately 2.4 eV for the (100) surface of the copper model used.

Therefore the following modifications to the earlier repulsive potential model were made. First, the top two layers of the sputtered surface were defined in a relaxed position, and second, an atom was considered sputtered only if it passed the surface with a "perpendicular" energy greater than 2.4 eV.

B. PRELIMINARY OBSERVATION OF FOCUSED COLLISION SEQUENCES.

Harrison [10] has shown, for argon ions incident normally on a (110) copper surface, that hard collisions can, for ion impacts on certain regions of the surface, occur relatively deep inside

the crystal. In these regions (see Fig. I.a.) incoming ions "channel" only in the sense that they are contained in the (110) channel described by Lehmann and Leibfried [17] as they move through the lattice. Instead of leaving the lattice undisturbed, however, they make occasional relatively hard collisions with atoms surrounding the channel. These events, according to Harrison, are the only ones which could initiate focused collision sequences back toward the surface.

For convenience in this paper, ions are said to channel if they simply satisfy the criterion of containment in the channel. Thus in Fig. 1.a., ions impacting in Region 1 channel with little interaction with the lattice, ions impacting in Region 2 channel with relatively hard interactions, and ions impacting in Region 3 do not channel.

A series of computer runs were made with 20 keV argon ions impacting in Region 2 on a lattice defined in the (110) surface configuration. Runs were made with the incoming ions moving in both normal and slightly off normal directions. The latter orientation was observed to cause primary collisions with channel atoms of greater frequency and magnitude as the ions "skimmed" [10] down the channel. The collisions invariably caused channel atoms to move off in directions nearly parallel to the surface.

For monocrystals of a practical size for repeated runs (about 400 atoms), collision cascades, initiated more than a few atomic layers deep, came out the sides of the crystal without reaching

the surface. Many atoms were observed to have substantial velocity components directed toward the surface, but it was impossible to determine if they did in fact cause sputtering. Therefore, in order to observe sputtering relatively far away from the impact point, several computer runs were made with ions impacting on one corner of the surface (see Fig. 1.b.). Many collision sequences were observed propagating along close packed rows of atoms, and a few were directed toward the surface and resulted in the sputtering of a surface atom. An example of such an event is shown in Figs. 1 and 2. Obviously this is a focuson in the sense described by Nelson and Thompson [4]. This event was initiated by a primary collision imparting 155 eV to a copper atom in the sixth atomic layer below the surface. It was impossible to observe sputtering from collisions much deeper than this without using a larger lattice than computer running time would tolerate.

C. COLLISION SEQUENCES INITIATED BY CHANNELED IONS

It was quite obvious, after the preliminary investigation just described, that a detailed determination of the overall significance of deep collisions in sputtering was impossible with techniques used in previous works. This is especially true for high ion beam energies. Therefore a two-step analysis, one of primary (ion-atom) collisions and a separate one of secondary (atom-atom) collisions, was undertaken. This analysis was applied to 20 keV argon ions incident obliquely on a (100) copper surface, with attention given primarily to ions contained in the ⟨110⟩ channels.

1. Primary Collisions in the ⟨110⟩ Channel

The object of this step in the investigation was to determine the initial kinetic energies and directions of motion of copper atoms after interaction with an incident ion passing down an adjacent channel. A modification to the computer program was made so that it defined a lattice which included only the atoms immediately adjacent to a ⟨110⟩ channel. It was determined by direct comparison that there was no significant difference between collisions in this model and those in the complete lattice when the ion channeled. The net result was a great reduction in program running time. Actual production computer runs contained the lattice defined to a depth of twenty atomic layers. The output contained the initial energy and direction of all atoms recoiling from collisions in the channel.

A set of impact points (see Fig. 3) was run for incidence angles of 29, 33, 37, 41, 45, 49, 53, 57, and 61 degrees from surface normal. The beam axis was rotated about a ⟨010⟩ direction in the surface in order to take advantage of the symmetry of the crystal to reduce the number of impact points which generates a complete set. Some important features of the results of this analysis are represented in Figs. 4 through 12. The representative areas in the figures are divided into six regions. Region 1 represents impact points where ions channel with collisions resulting in less than 200 eV energy transfer per collision, Region 2 represents less than 500 eV energy transfers, Region 3 represents less than

1 keV, and Region 4 represents greater than 1 keV energy transfers. Ions were considered to be channeled if they were contained in the channel to a depth of four or more atomic layers. In Region 5 the incoming ions did not channel but deposited their energy locally about the impact point. Region 6 represents an area where energy was deposited locally, but the resultant sputtering was markedly less than in Region 5. This was due to the following two effects. Either the incoming ion reflected off the top atomic layer carrying away a significant portion of its incident energy, or it underwent an extremely hard surface interaction which initially sputtered a single top layer atom imparting to it several keV of energy.

It was also noted that a number of incoming ions actually entered the $\langle 110 \rangle$ channel but after penetrating a few atomic layers escaped without a hard collision with one or more of the channel atoms. These impact points are marked on Figs. 4 through 12 by an "X". Further some of the ions interacted with a surface atom before entering and subsequently escaping the channel. These impact points are marked by a circled "X" ($\otimes$).

2. Secondary Collisions

One would expect that the major portion of the sputtering from primary collisions occuring several layers deep is due to recoil atoms of substantial energy which initially have velocity components directed toward the surface. It is this type of primary recoil which is of major interest in this part of the analysis.

Preliminary computer runs indicated that primary recoil atoms with less than 200 eV seldom caused sputtering when initiated

more than a few layers below the surface. Also it was noted in the results of the primary collision analysis that a few recoil atoms had energies of more than 1 keV with large velocity components directed toward the surface. Production computer runs were subsequently made with primary recoil energies of 200 eV, 500 eV, 1 keV, and 2 keV with an isotropic spread of recoil directions (see Fig. 13) represented by points in the upper half of a sphere about a lattice site. Because of the symmetry of the crystal, it was only necessary to run one-eighth of a total set of such directions. The total set may be generated by repeated reflections of the reduced set shown. Data is represented by projecting the reduced set from the sphere about the lattice site to a curved triangular surface (see Figs. 14 through 17). The numbers on these projections represent the number of atoms sputtered. In making the counts of sputtered atoms, those which only marginally met the sputtering criterion previously specified were included. Therefore the numbers are to be considered as somewhat optimistic. A figure is included for the specified recoil energies showing sputtering from depths of four, eight, and twelve atomic layers.

3. Sputtering Estimates

By comparing the energy and direction of motion of each recoil atom to the figures just described, a reasonable estimate of the number of atoms sputtered by each primary collision recoil was obtained. Only recoil atoms moving toward the surface were

considered to contribute. By considering all such primary recoils for each incident ion, it was possible to obtain an averaged value for the "partial" sputtering ratio associated with each of Regions 1 through 4 in Figs. 4 through 12. By comparing these values to their respective areas on the figures, an estimate of the sputtering due to deep channel collisions was obtained for the specified angle of incidence. The ratio of the area of a particular region to the total area of the representative area is the fraction of the total beam cross-section which causes the type of collision the region represents. The results are given in Table I as are experimental sputtering ratios due to Fluit and Rol [6].

In Regions 5 and 6 in Figs. 4 through 12, all significant primary collisions occur in a random fashion near the impact point. However it was noted, for 20 keV impacts in these regions, that a number of high energy secondary recoil atoms came out the sides of the defined lattice. These atoms typically carried away several keV of energy. Thus it was not certain that all significant sputtering events were contained in the lattice used. However it was possible to make a few multiple runs which effectively contained all significant collisions. First a single run was made which contained all primary interactions and then additional runs were made as needed to contain the secondary recoil atoms which came out the sides of the first lattice. The excessive computer time required limited this type of investigation to only a few typical

impact points. A rough estimate of the number of atoms sputtered from impact points in Regions 5 and 6 were obtained in this manner, thereby making it possible to make an estimate of the total sputtering ratio. These are also shown in Table I.

IV. RESULTS

A. SECONDARY COLLISION ANALYSIS

The most striking feature of the data in Figs. 14 through 17 is the large, almost random differences in the number of atoms sputtered for the different recoil directions investigated. Although not of particular interest here, there was a similarly striking random variation in the energies of the sputtered atoms. This clearly indicates that the predominant sputtering mechanism in this case is a random cascade rather than focused collision sequences.

Also the gross behavior, as indicated by the averaged sputtering values shown in the figures, must be noted. 200 eV primary collisions at 12 atomic layers below the surface produce no sputtering, and even 2 keV collisions show a marked reduction in sputtering when occuring at this depth. Thus it appears that focused collision sequences are damped rather quickly. A good example showing the magnitude of such damping is illustrated in Fig. 2. This is not to say that focusing effects do not contribute to sputtering, but simply that the data indicates that random collisions dominate the effect.

B. PRIMARY COLLISION ANALYSIS

On the basis of the secondary collision data, it can be said that Figs. 4 through 7 represent fairly well the "transparency" (in a similar sense to that suggested by Fluit and Rol) of the

representative area with respect to the ⟨110⟩ channel at the specified angle of incidence. Region 1 is the most transparent and Region 5 is the least as far as penetration into the ⟨110⟩ channel is concerned. Quite obviously Region 6 must be considered separately. Region 1 is largest in area for incidence angles of 41 and 45 degrees. This corresponds rather well to the minimum in an experimental curve [6] which corresponds approximately to the ⟨110⟩ direction. The fact that this minimum does not in either case occur at exactly 45 degrees can be explained by a slight deflection (scattering) of the incoming ions by the relaxed surface atoms.

The presence of Region 6 is evidently significant, especially for large incidence angles. A number of computer runs, using a complete lattice, were made with ions impacting in this region which indicates that the sputtering is on an average about 50 percent less than in Region 5.

The areas defined by "X's", while representing an effect beyond the scope of this work, do suggest an interesting aspect of transparency. It would seem that channels associated with low index crystal directions overlap more than would be expected if a hard sphere model [6] were considered. As the circled "X's" (⊗) in the figures indicate, this is due to scattering by surface atoms of the incoming ions into channels other than the one corresponding to the beam incidence angle. This effect could possibly explain the relative width and position of the minima in the

sputtering ratio versus incidence curves due to Fluit and Rol. Because of its lower mass, a 20 keV neon ion is scattered more readily than an argon. Thus, in addition to an overall reduction in sputtering, one would expect to find the minima corresponding to low index channels less well defined. This is certainly the case in the data of Fluit and Rol.

C. SPUTTERING ESTIMATES

From consideration of primary and secondary collision data, it was determined that ions impacting in Region 1 in Figs. 4 through 12 produce essentially no sputtering, ions impacting in Region 2 sputter about one atom per ion, in Region 3 ions sputter about three atoms per ion, and in Region 4 about six atoms per ion. Using this criterion (see Table I) one can attribute about 25 percent of the sputtering to collisions relatively deep in the crystal at an incidence angle of 29 degrees, and about ten percent at 45 degrees where the ⟨110⟩ channel is well defined. However, sputtering due to both deep collisions and surface effects are included in the numbers of sputtered atoms per ion given above. Therefore these estimates must be considered high.

Also from consideration of a number of computer runs using a complete lattice, it was determined that ions impacting in Region 5 sputter about 15 atoms, and in Region 6 about eight atoms. Using this and the above channel collision criterion, the total sputtering ratios shown in Table I were obtained. Taking into account the rather gross approximation made to obtain these results, the

numerical values should not be taken too seriously. However, the fact that the sputtering ratios obtained have roughly the same values as those obtained experimentally indicates that Figs. 4 through 12 and the interpretation given to them are approximately correct. Clearly, one could adjust the values given the "partial" sputtering ratios in each of Regions 1 through 6 and obtain a much better fit, but this was not the point of the exercise.

This approach predicts a rather sharp increase in the sputtering ratio for low (greater than 45 degrees) beam incidence angles (these data were not given by Fluit and Rol). This is to be expected, at least to a point, since incoming ions will initiate cascades for the most part in the surface atomic layers. However, at some point, ion reflection off surface atoms will become the dominant factor and the sputtering ratio will drop off rapidly. No precise information about the sputtering ratio versus incidence angle curve can be obtained in this region from the data as presented, since no quantitative interpretation can be given to the regions marked by "X's". Presumably these areas are low index channels other than the $\langle 110 \rangle$.

V. CONCLUSION

The results of this investigation may be summarized as follows:

(1) The primary collision analysis strongly supports transparency as an important consideration in sputtering from single crystals.

(2) The secondary collision analysis indicates that random collision cascades are the predominant mechanism in producing sputtering from primary collisions deep within a lattice.

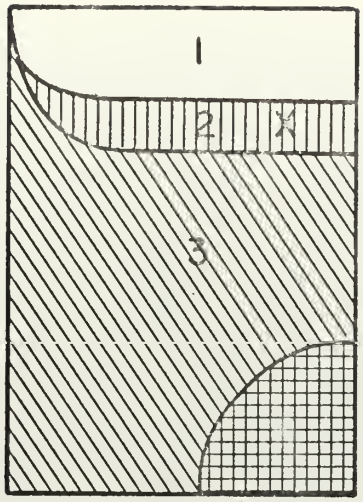
(3) Deep primary collisions account for up to 25 percent of the total sputtering from a single crystal, depending upon the angle of incidence. However only a small fraction of this can be attributed to focused collision sequences for reasons already given.

(4) Primary ion reflection off surface layers is a significant factor in sputtering from oblique angle ion beam incidence when the ion mass is less than the atomic mass.

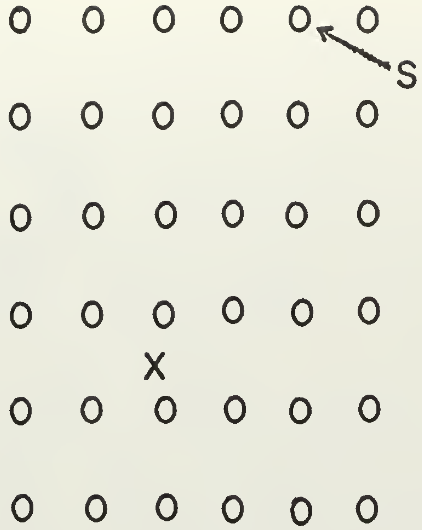
(5) The somewhat puzzling variations in structure of sputtering ratio versus beam incidence curves due to different ion species and energies may be explained by low angle scattering of incoming ions off surface atoms.

Therefore sputtering is, in general, a complicated combination of channeling with subsequent deep primary collisions, hard primary collisions at or near the surface, primary reflection from surface layers, and effects of initial ion interations with surface

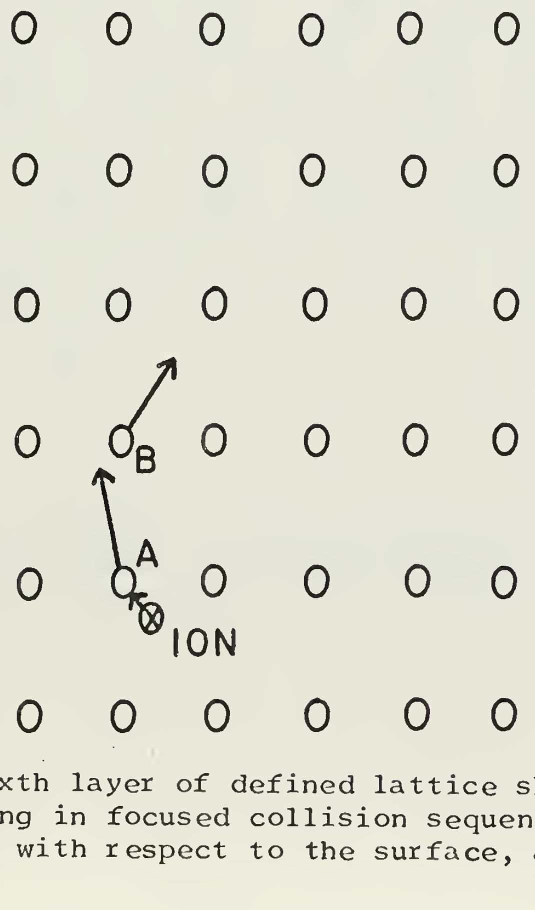
atoms. The contribution of primary ion reflection must be carefully studied before the relative importance of all of the mechanisms can be completely understood.



<u>Figure 1.a.</u> Representative area of (110) surface of copper (according to Harrison [10]). Impact point of 20 keV argon ion is marked by "X".



<u>Figure 1.b.</u> Surface of lattice defined in computer for this example. "X" is the ion impact point. Atom "S" was sputtered by focused collision sequence.



<u>Figure 1.c.</u> Sixth layer of defined lattice showing primary impact resulting in focused collision sequence. Atom "A" moved downward with respect to the surface, and atom "B" moved upward.

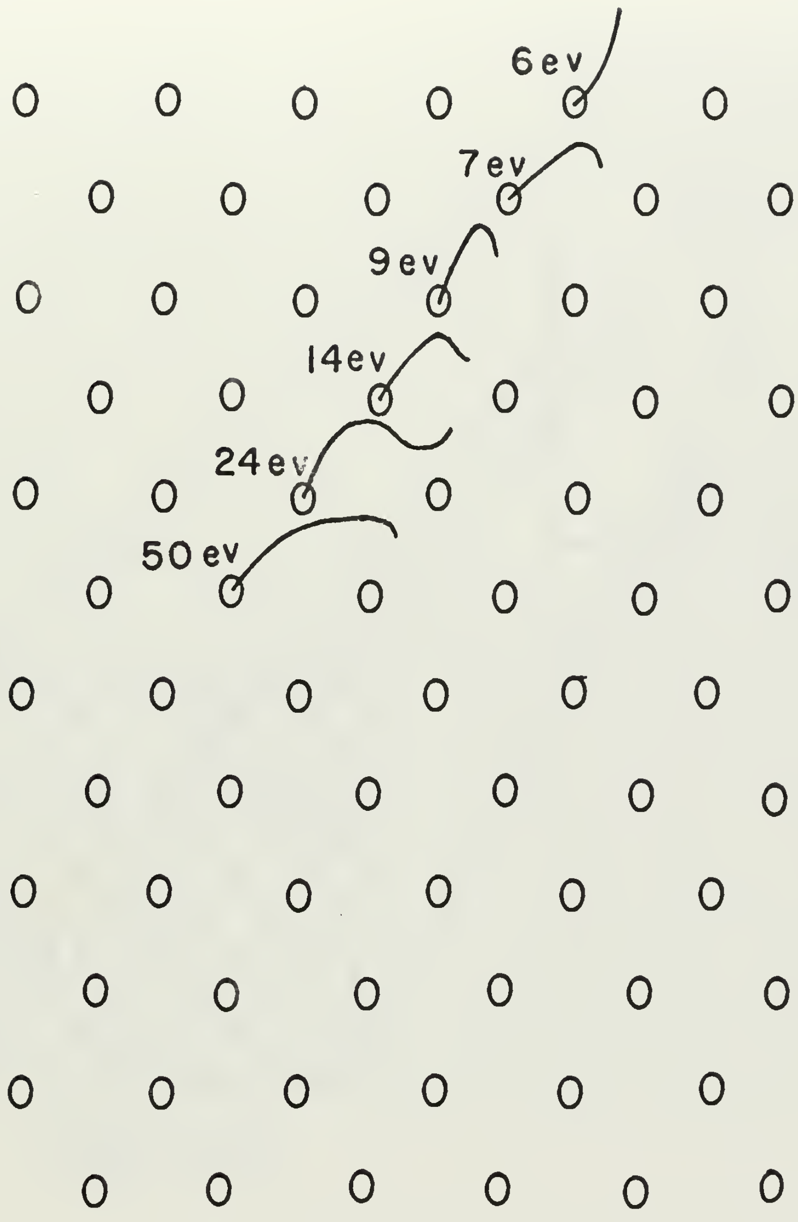


Figure 2. (111) planar section of defined lattice showing paths of atoms in focused collision sequence. Initial energies imparted to each atom are also shown.

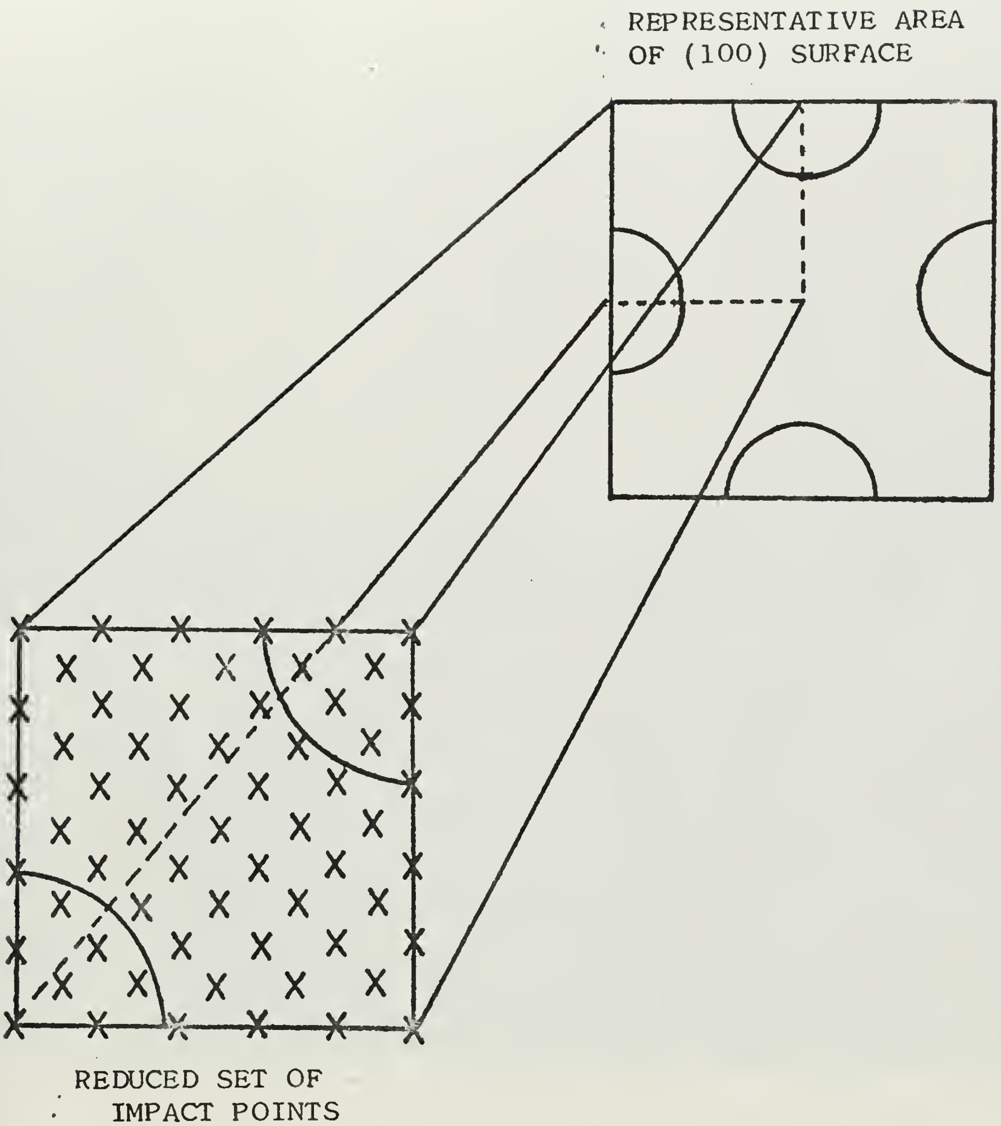


Figure 3. Ion impact points run on computer for oblique ion incidence.

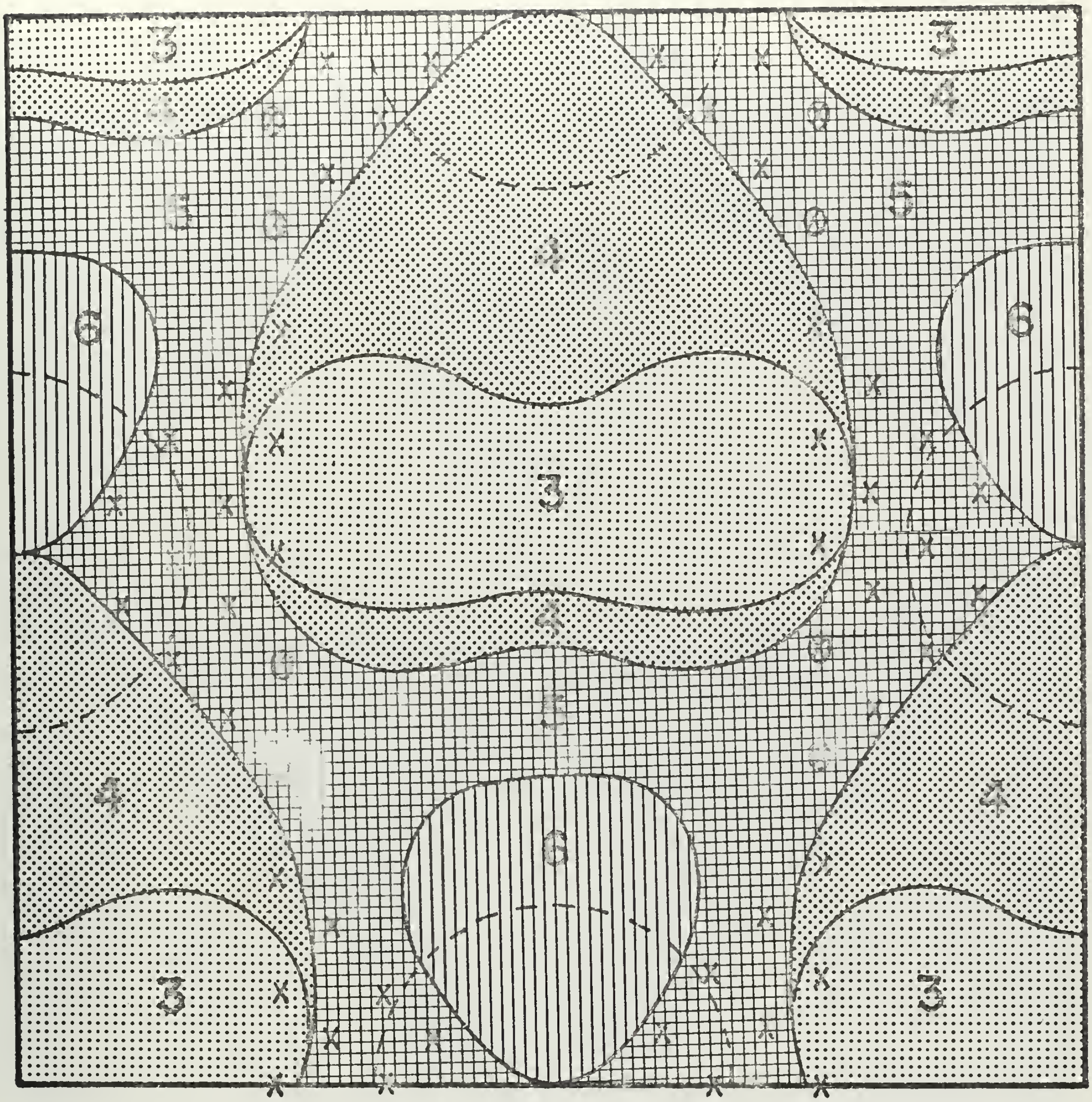


Figure 4. Representative area for 29 degreee ion incidence. Dotted circles represent atomic sites in surface layer of crystal. The significance of the regions numbered 1 through 6 and the symbols "X" and "ⓧ" are discussed in the text of this paper.

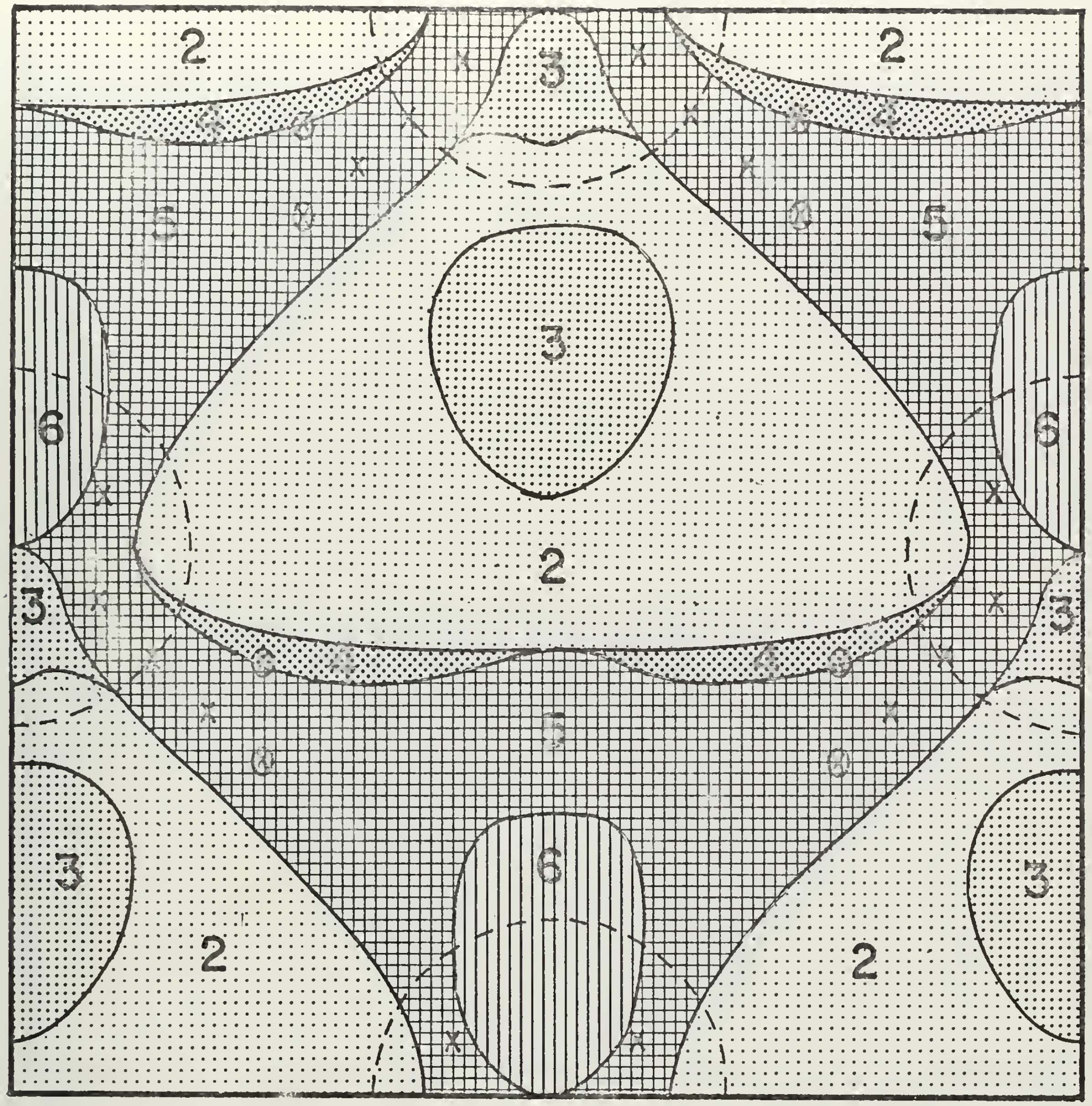


Figure 5. Representative area for 33 degree ion incidence.

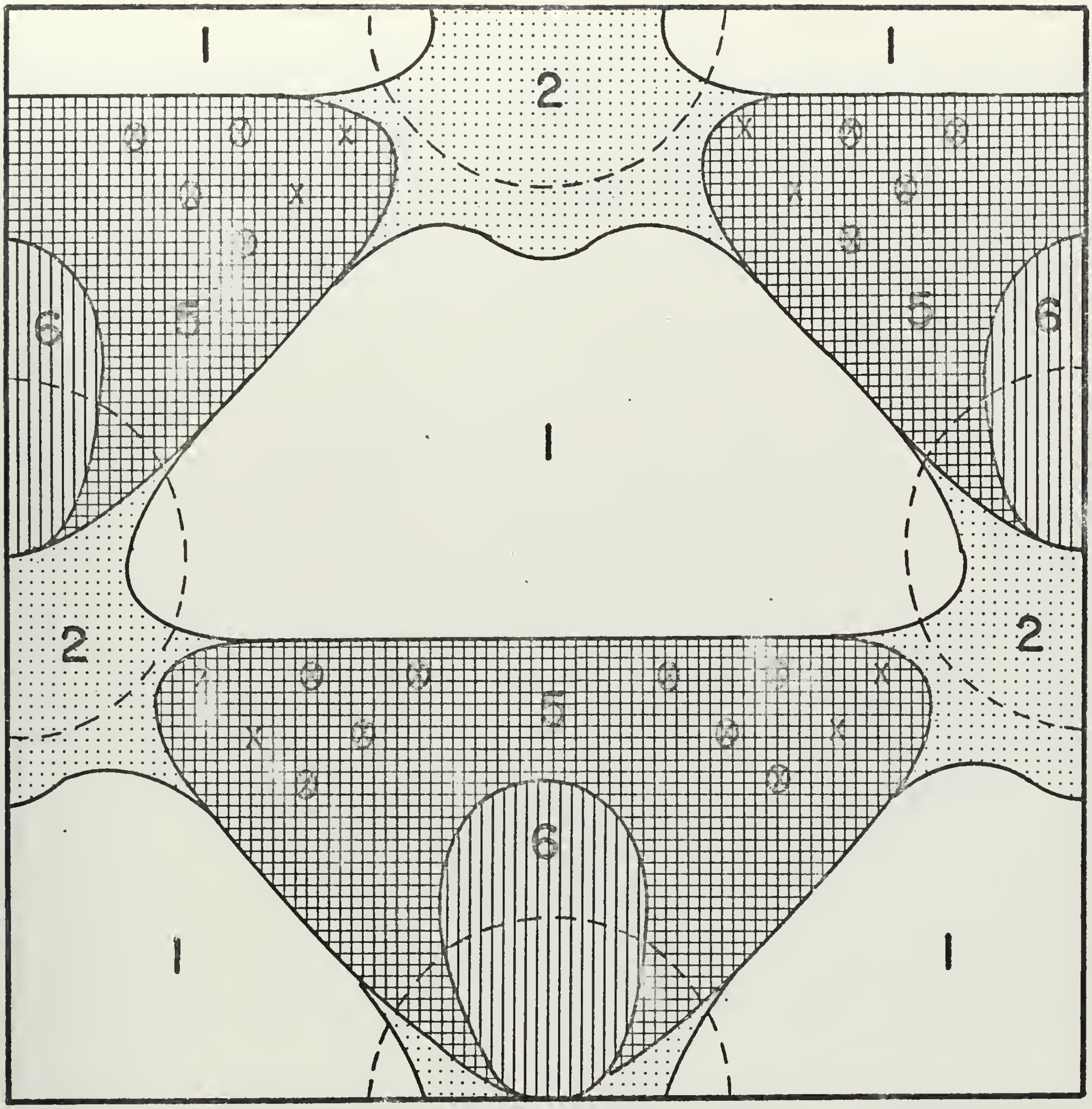


Figure 6. Representative area for 37 degree ion incidence.

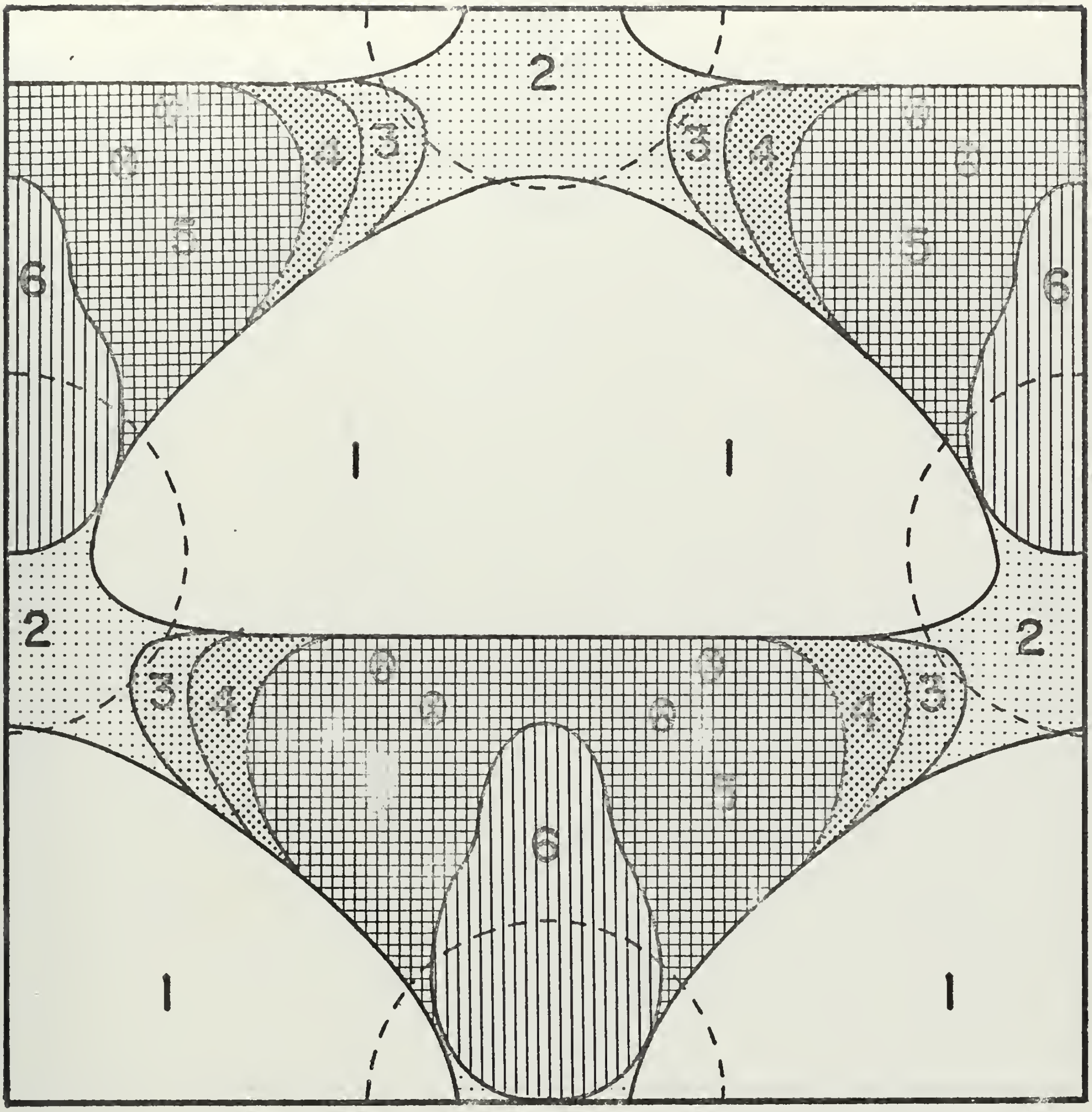


Figure 7. Representative area for 41 degree ion incidence.

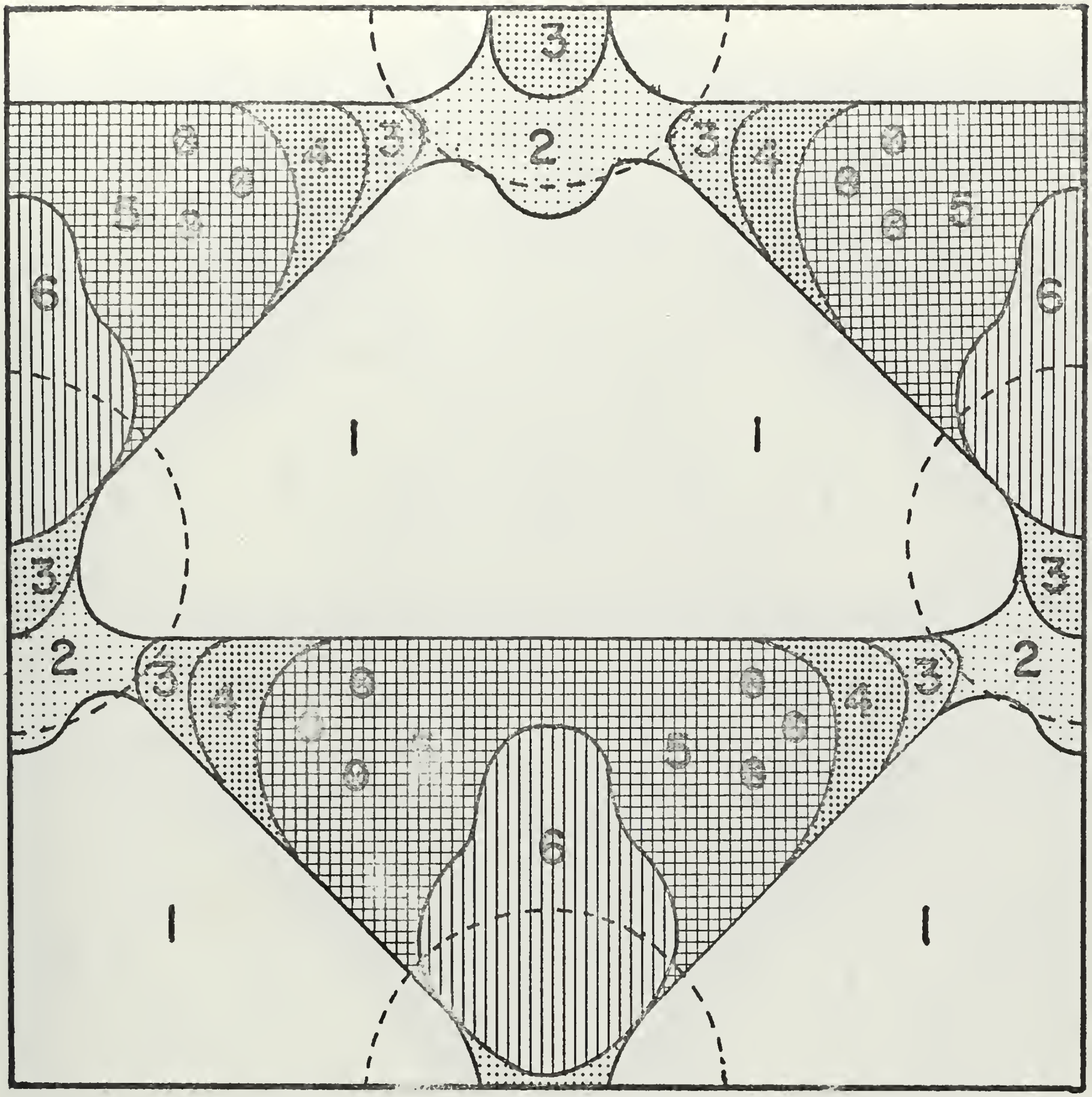


Figure 8. Representative area for 45 degree ion incidence.

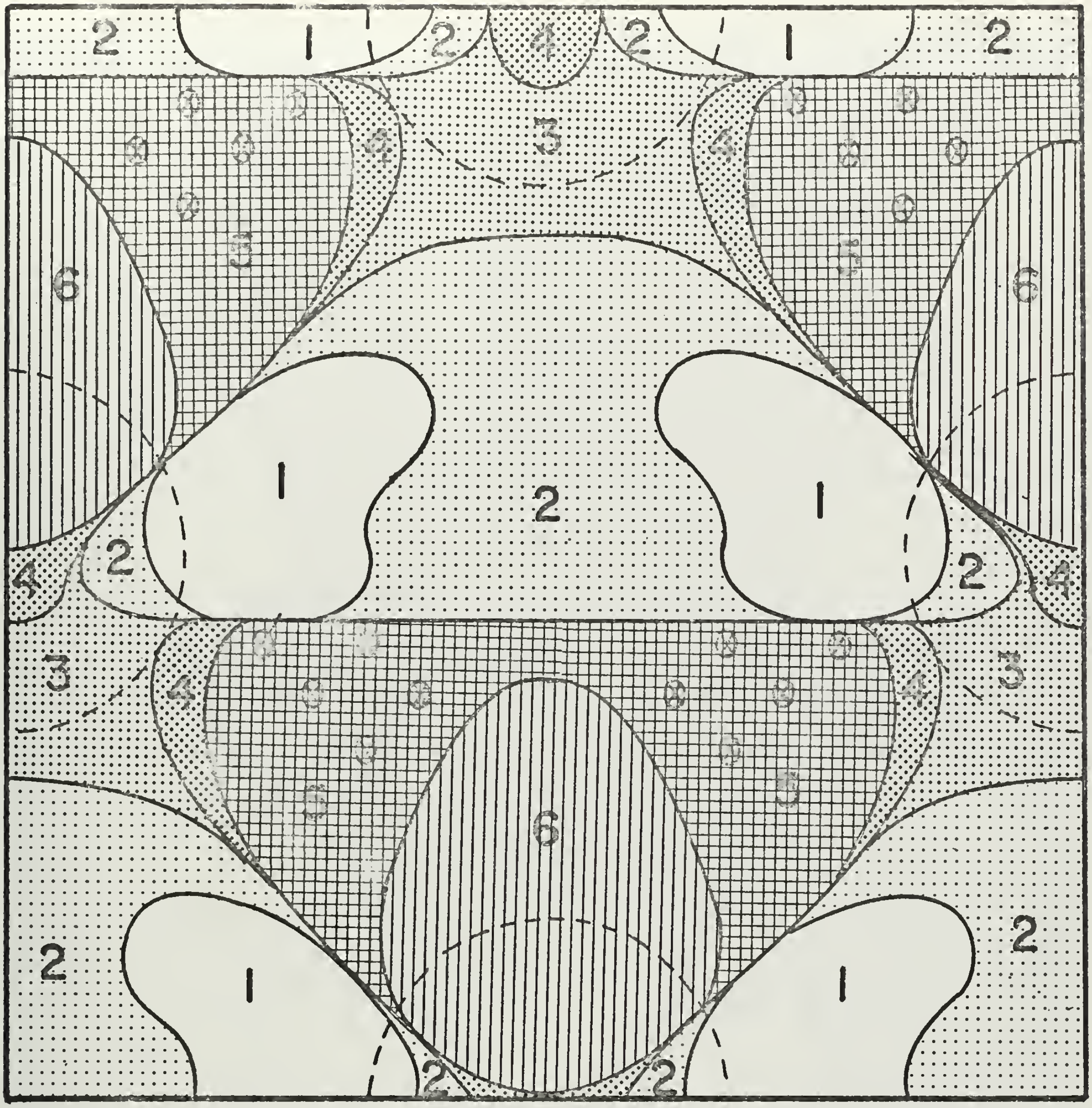


Figure 9. Representative area for 49 degree ion incidence.

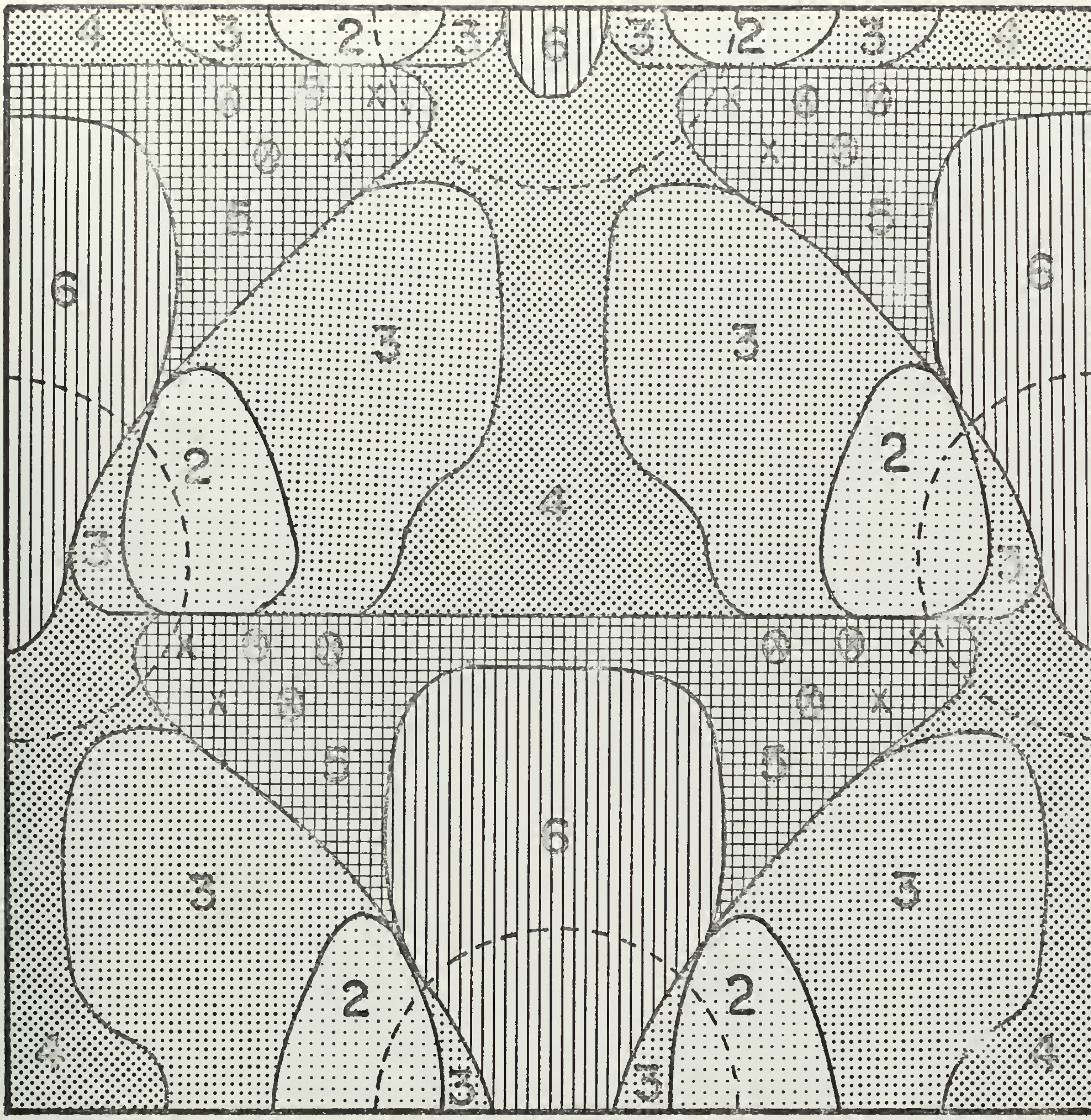


Figure 10. Representative area for 53 degree ion incidence.

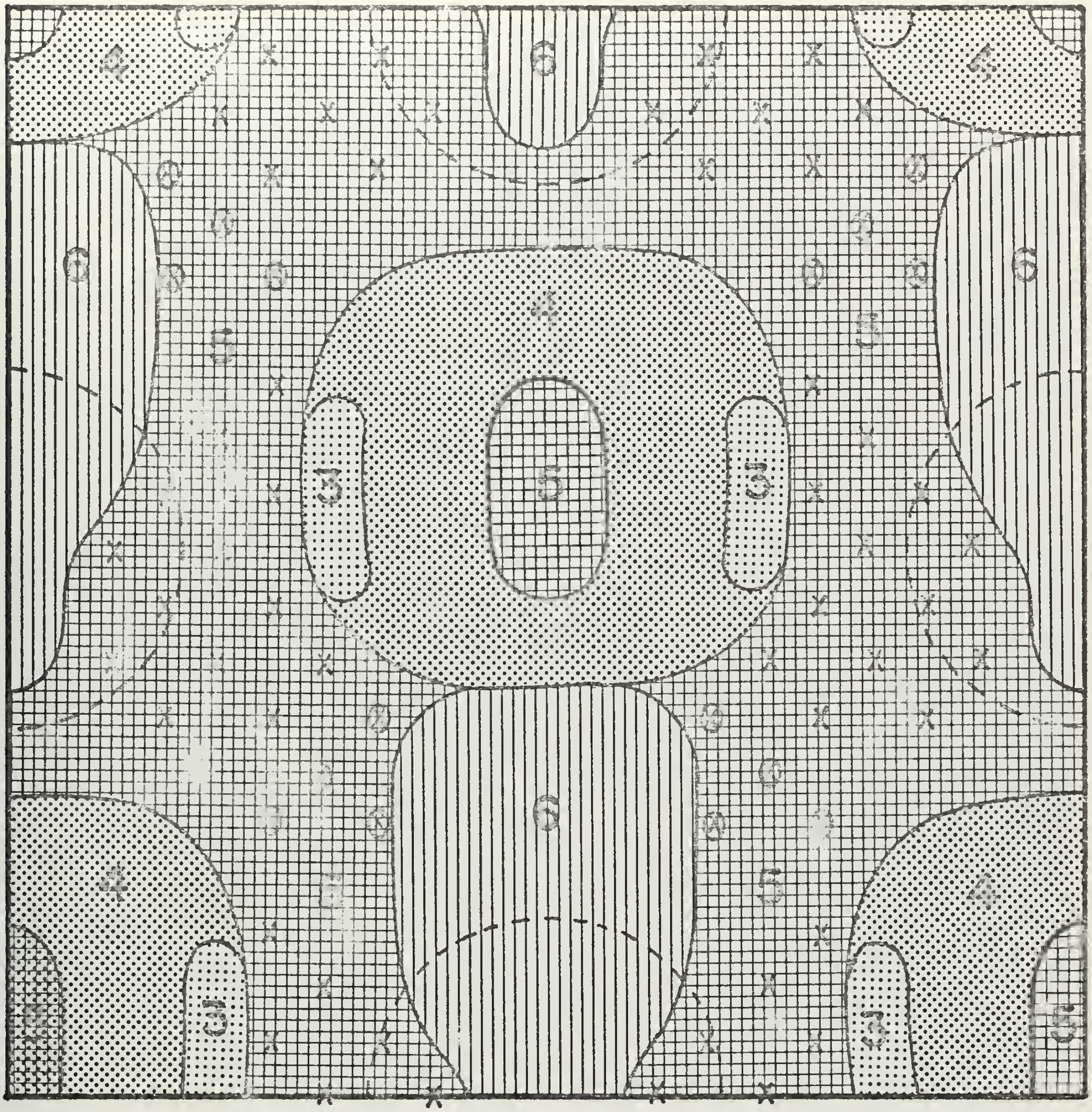


Figure 11. Representative area for 57 degree ion incidence.

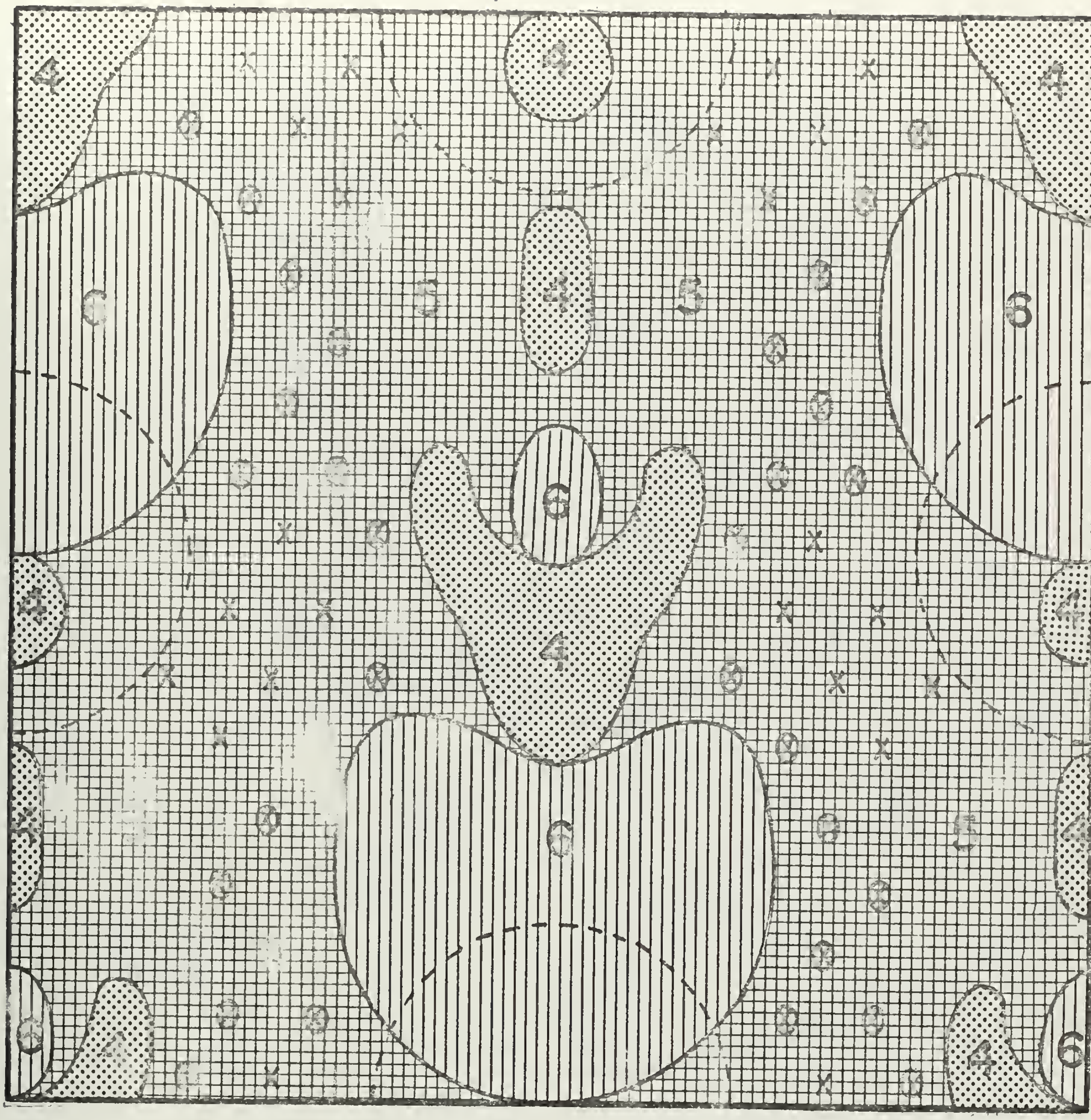
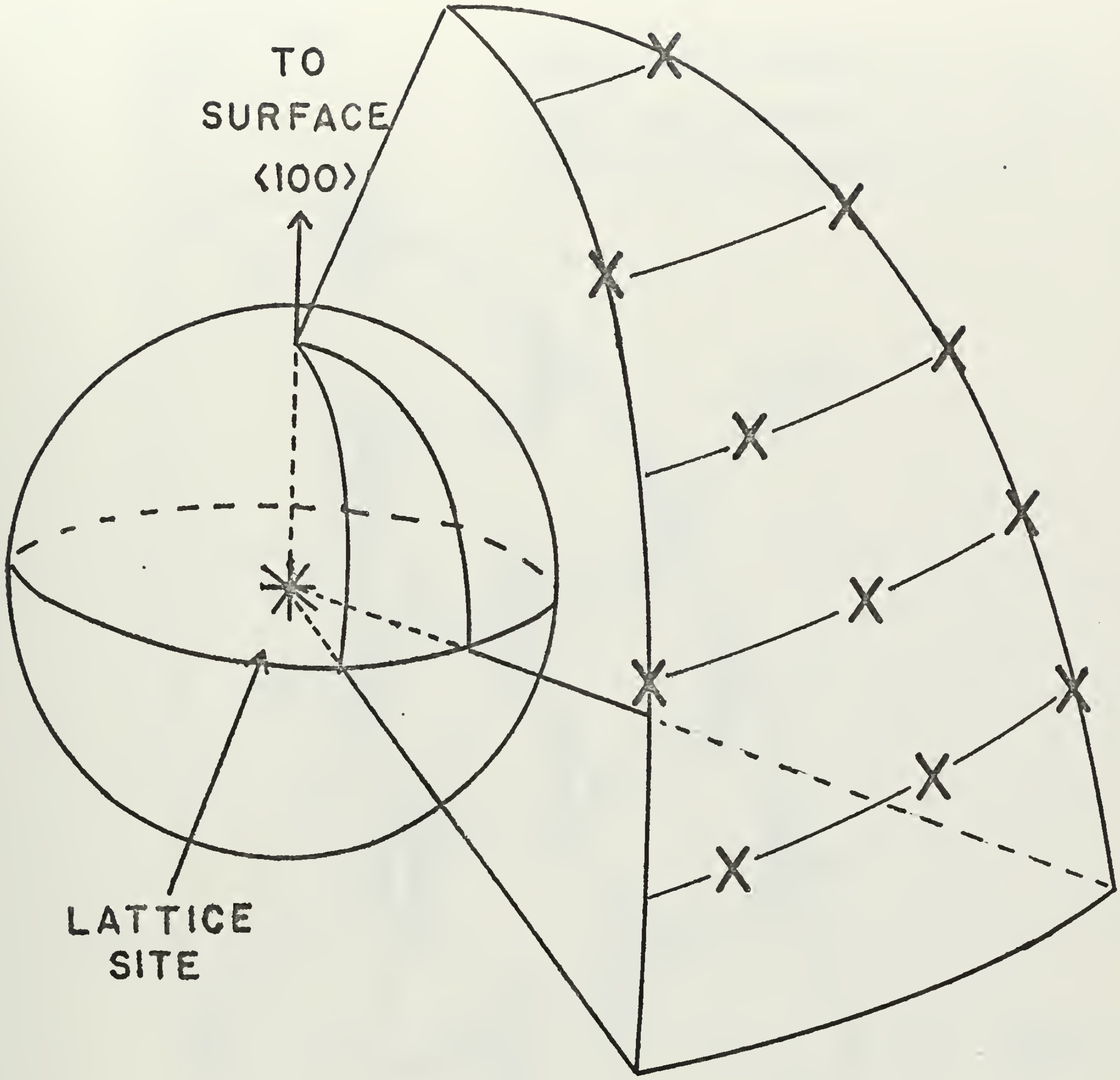


Figure 12. Representative area for 61 degree ion incidence.



<u>Figure 13.</u> Reduced set of primary recoil directions.

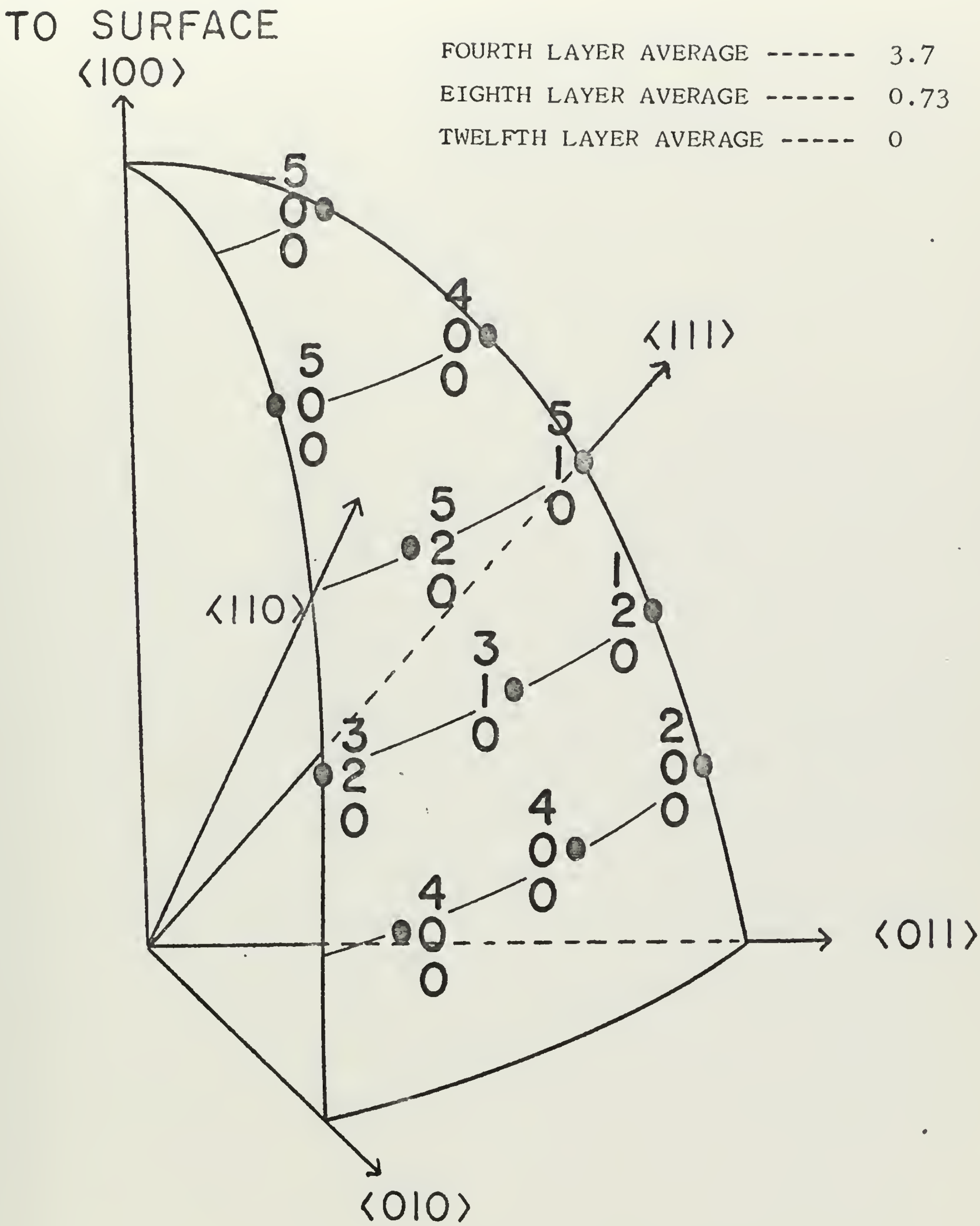


Figure 14. Sputtering from 200eV primary recoil atom. The upper number is the number of atoms sputtered from a primary recoil in the fourth atomic layer. The middle number is from the eighth atomic layer, and the bottom number is from the twelfth.

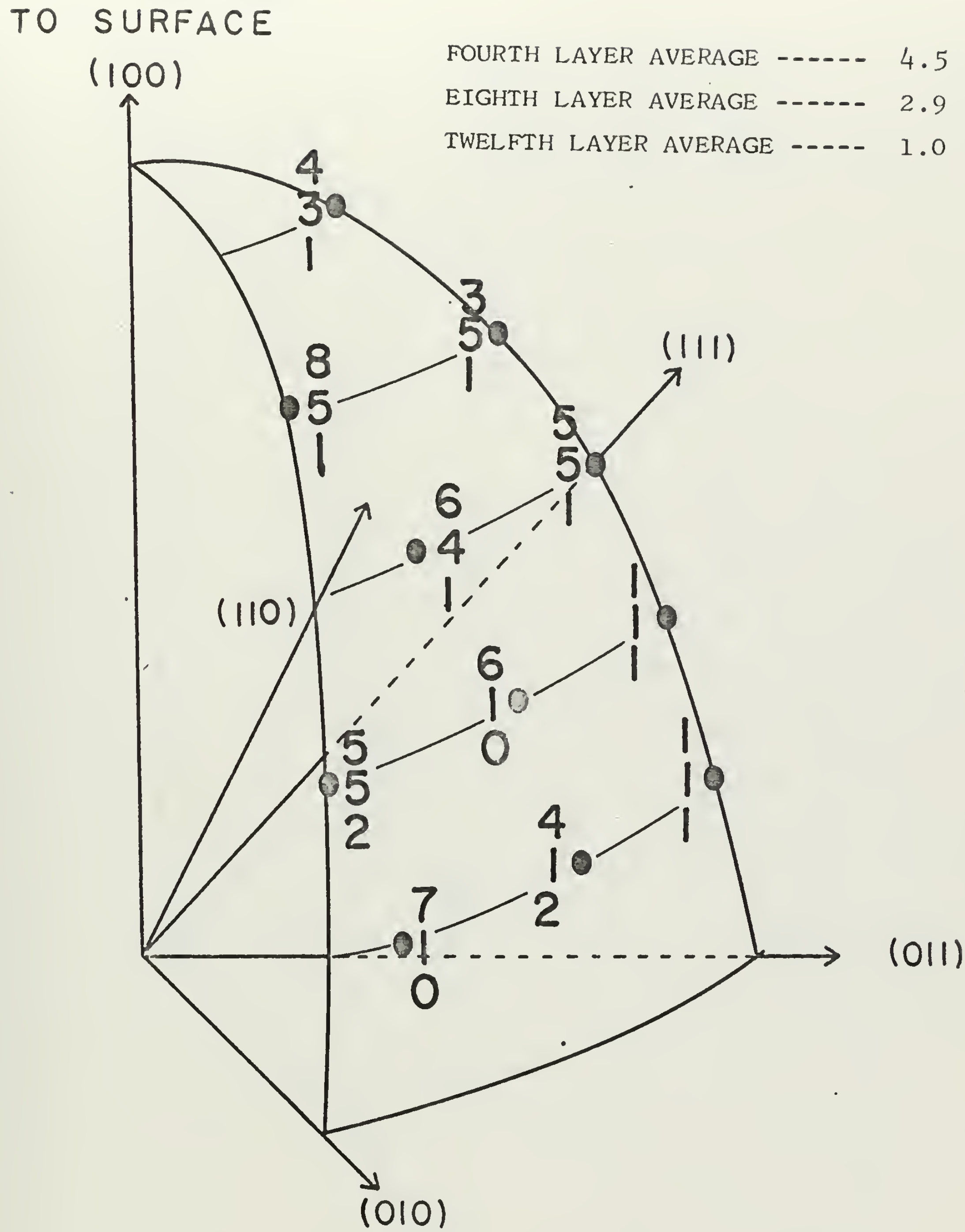
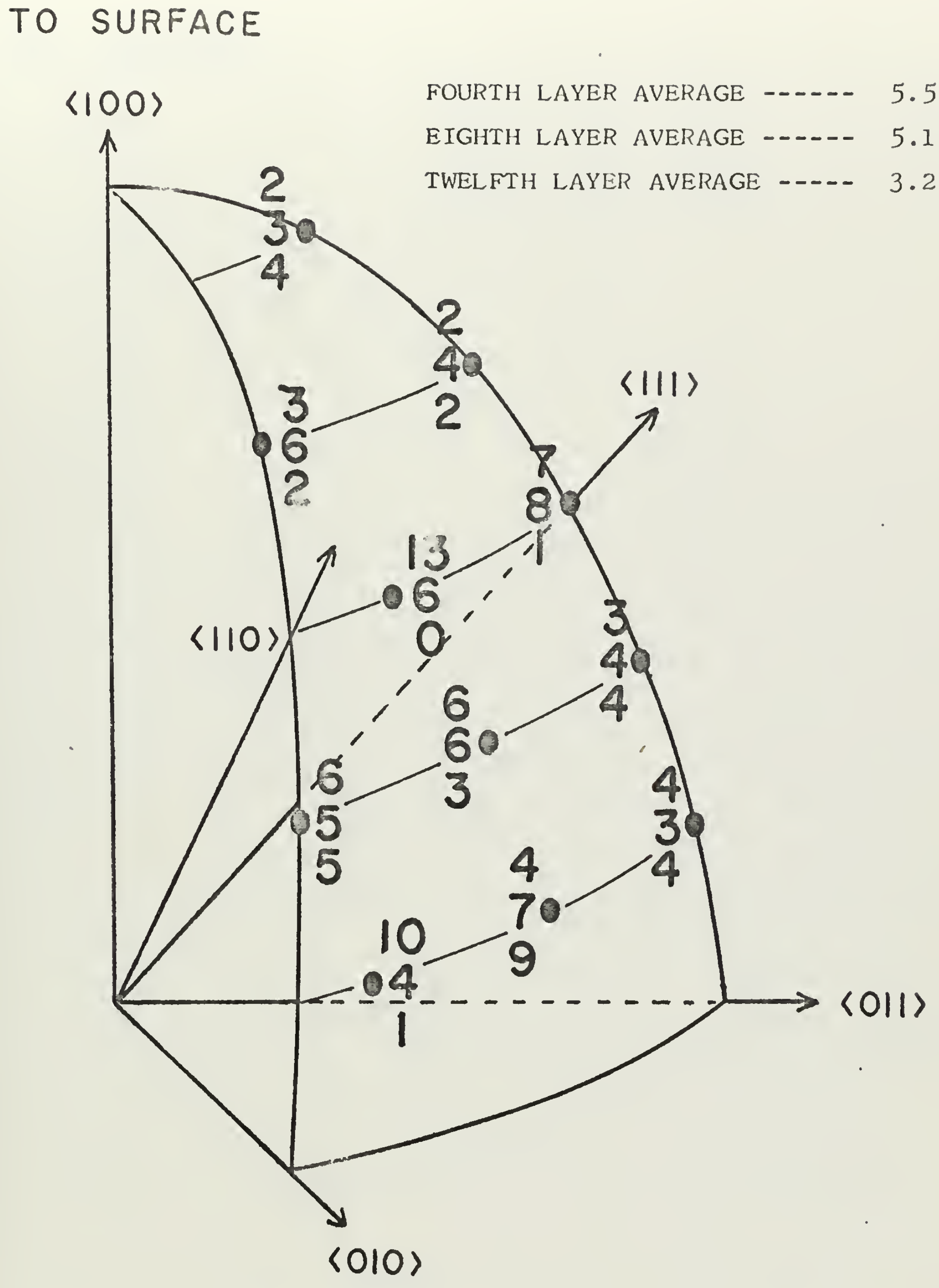


Figure 15. Sputtering from 500eV primary recoil atoms.



<u>Figure 16.</u> Sputtering from 1 keV primary recoil atoms.

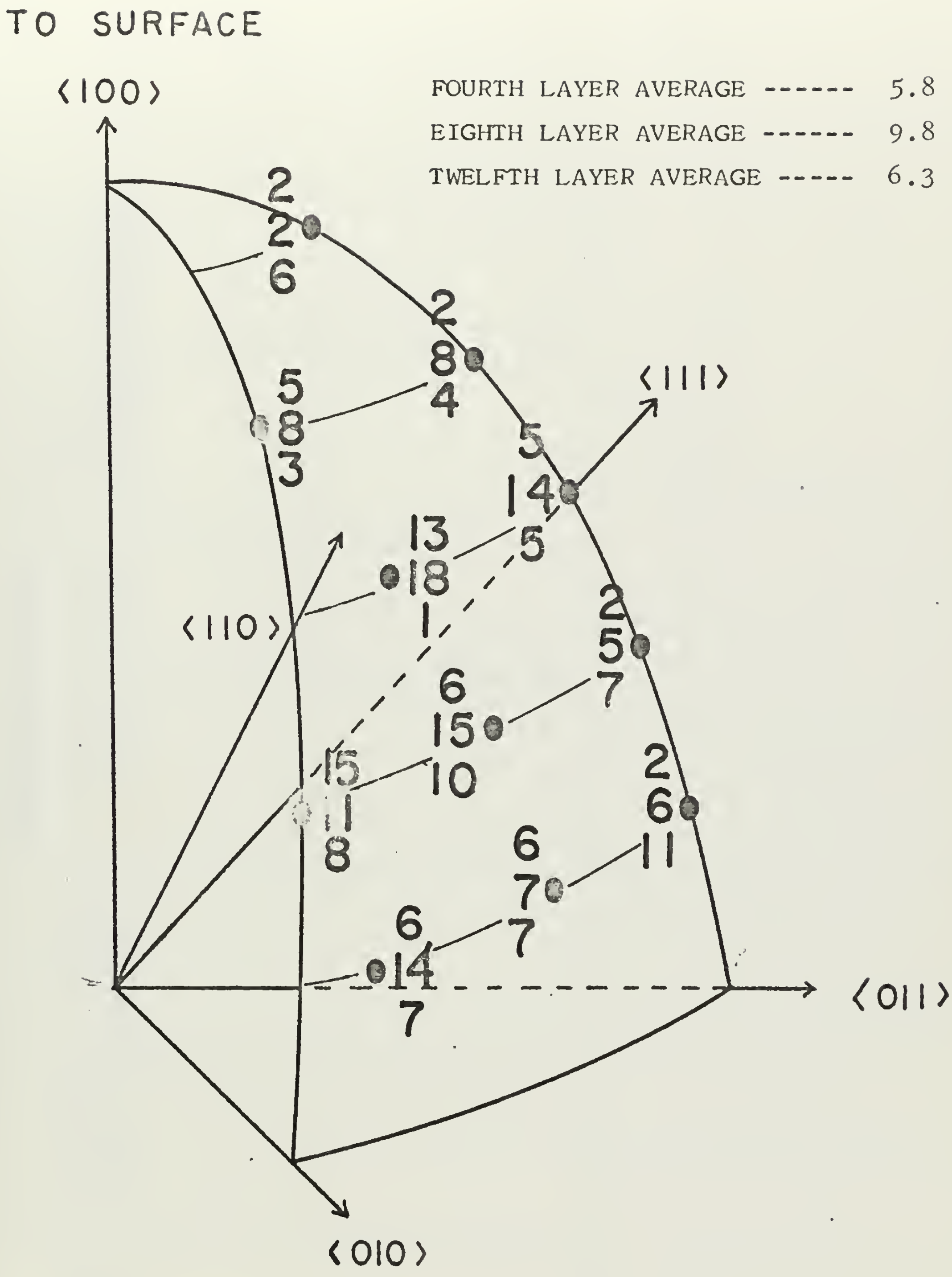


Figure 17. Sputtering from 2 keV primary recoil atoms.

Table I. Sputtering Estimates

	SPUTTERING RATIOS		
ANGLE OF BEAM INCIDENCE	EXPERIMENTAL RESULTS (FLUIT AND ROL [6])	RESULTS OF THIS INVESTIGATION	PARTIAL RATIO DUE TO DEEP COLLISIONS
29	9.5	9.0	2.3
33	6.7	6.8	0.96
37	4.5	5.5	0.15
41	3.1	4.0	0.38
45	3.5	4.2	0.35
49	5.3	4.8	0.76
53	---	6.2	1.7
57	---	10.8	1.8
61	---	13.2	0.64

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13. ABSTRACT

A self-contained computer simulation of sputtering from 20 keV incident ions is not possible with present computers. However, a simulation can be done by considering primary and secondary collisions separately. An investigation of 20 keV argon ions incident obliquely on the (100) surface of a face-centered cubic copper crystal was done at angles from 29 to 61 degrees from normal. Results strongly support the concept of transparency, but indicate that focused collision sequences make very limited contributions to sputtering. Depending on the ion beam incidence angle, up to 25 percent of the sputtering may be due to random collision cascades initiated by deep primary collisions. The remainder is caused by surface collision mechanisms. Reflection of incident ions off surface atoms significantly affects argon-copper sputtering when the ions are obliquely incident.

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